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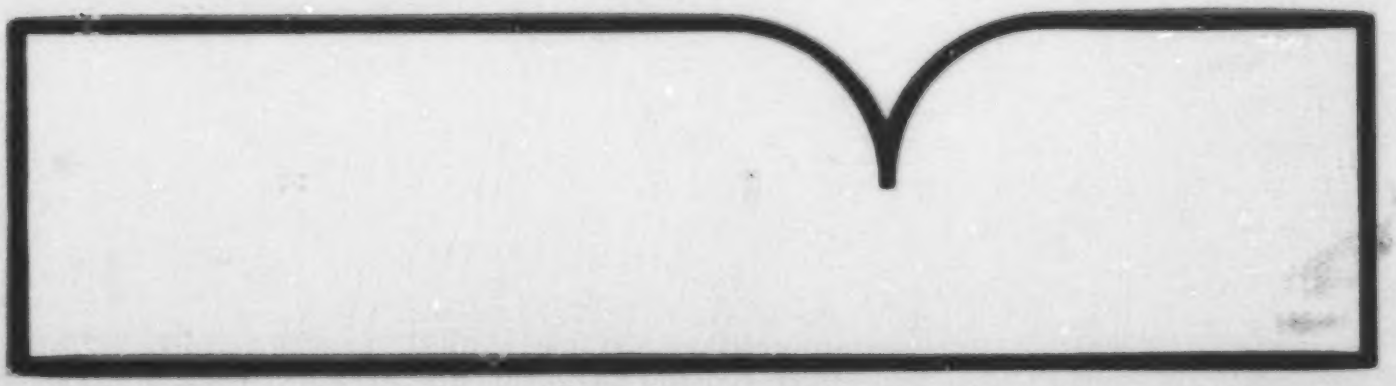

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REGIONAL ANALYSIS OF WET DEPOSITION FOR EFFECTS RESEARCH

by

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16. ABSTRACT

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Important issues in data preprocessing include temporal resolution, data stratification into geographic regions, and choosing between direct and indirect methods for interpolating wet deposition. The "indirect method" involves interpolating precipitation amount and concentration separately and using their product for wet deposition maps. The indirect method is recommended because it allows the use of more spatially dense precipitation amount data sets, thereby better representing variability in the precipitation process. Limited experimental evidence does not demonstrate spatial correlation between that precipitation amount and concentration, a condition for the use of the indirect method. It is recommended that further investigation of the degree of independence of precipitation amount and concentration across space be preformed.

Given that there are many ways to weight near and distant data for estimating data at a non-monitored site, available spatial interpolation techniques are discussed. The geostatistical technique, kriging, is discussed in detail to allow other researchers the benefit of previous applications to precipitation chemistry. Different interpolation techniques may produce maps that are similar but estimation variances that are different, or absent. Procedures for generating and checking uncertainty estimates are discussed.

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Abstract

The basis for regional analysis of precipitation amount, concentration, and deposition is investigated. When performing such a spatial analysis, key issues are the data selection, data compositing, the interpolation technique, and the uncertainty of the results. Sources of data on precipitation amount and chemical composition are presented along with procedures for screening the chemical data. A review of recent work reveals that different scientists select different data sets and that data selection plays an important role in the resulting maps.

Important issues in data preprocessing include temporal resolution, data stratification into geographic regions, and choosing between direct and indirect methods for interpolating wet deposition. The "indirect method" involves interpolating precipitation amount and concentration separately and using their product for wet deposition maps. The indirect method is recommended because it allows the use of more spatially dense precipitation amount data sets, thereby better representing the variability in the precipitation process. Limited experimental evidence does not demonstrate spatial correlation between that precipitation amount and concentration, a condition for the use of the indirect method. It is recommended that further investigation of the degree of independence of precipitation amount and concentration across space be performed.

Given that there are many ways to weight near and distant data for estimating data at a non-monitored site, available spatial interpolation techniques are discussed. The geostatistical technique, kriging, is discussed in detail to allow other researchers the benefit of previous applications to precipitation chemistry. Different interpolation techniques may produce maps that are similar but estimation variances that are different, or absent. Procedures for generating and checking uncertainty estimates are discussed.

Preface

A workshop on regional analysis of wet deposition was held June 7-8, 1988 in Corvallis, Oregon. The coauthors of this report were participants in the workshop. In order to unify current work on spatial analysis some unpublished work by the coauthors has been referenced as a "workshop presentation". Any preference for certain data preprocessing or interpolation techniques expressed herein is based on the current practice of the workshop participants and available evidence from the 'acid deposition literature'. Techniques are expected to evolve with further research such that the conclusions of this report should be considered a snapshot in time.

1 Introduction

This report discusses issues associated with current efforts to interpolate and map precipitation chemistry data. Because of its usefulness for regional analysis, kriging is discussed in detail.

1.1 Rationale

Recent investigations have revealed that eastern U.S. forests are declining (McLaughlin,1985; Hornbeck and Smith,1985; Johnson and Siccama,1983) and some eastern lakes have become acidic (Landers et al.,1988; Wright,1988; Sullivan et al.,1988). One possible hypothesis is that acidic deposition represents a stress to these ecosystems that, when combined with natural stresses, has caused these changes. Woodman and Cowling (1987) suggested that three conditions must be satisfied to prove causation: 1) a mechanism, 2) a dose-response relationship, and 3) spatial and temporal consistency. To examine spatial consistency one constructs maps of potential stresses and damage. A gradient in a critical stress variable should correspond to a similar gradient in ecosystem response absent other contributing factors. If the gradients in the wet deposition of certain chemical species and forest damage (or lakewater acidity) are similar one requirement for a proof of causality has been met, namely "spatial consistency".

Maps, if constructed properly, are very useful for visualizing spatially oriented data. Regional estimates expressed as isopleth maps can reveal the magnitude and extent of acidic deposition and locate areas of high or low deposition. Consider a recent study of the contribution of a pollution source to downwind acidic deposition (Vong et al.,1988). The authors present their results both in the form of maps and as output from a statistical model. Which presentation of their data communicates the result most successfully ?

Spatial interpolation is necessary to generate maps of wet deposition and to estimate data for non-monitored locations. Interpolated values for wet deposition are used as inputs to aquatic and terrestrial ecosystems in the absence of measurements (Sullivan et al., 1988). This report reviews issues related to interpolating data and charting patterns in precipitation chemistry and wet deposition.

The first choices which must be made in performing a regional analysis of precipitation chemistry involve data selection. This process is more critical than generally has been acknowledged because the sources of chemical and precipitation amount data are numerous. If wet deposition is the desired regionalized characteristic, a consideration is whether to interpolate the data directly or to combine previously interpolated precipitation concentrations and amount. This choice forces an evaluation of the representitiveness of the various data sources and the spatial independence of concentration and precipitation amount. Finally a methodology must be selected for interpolating between the monitored data and estimating uncertainties.

The chemical species of likely interest to foresters and limnologists include those which control the acidity of the precipitation or induce/neutralize acidity in receiving systems such as the concentration and/or deposition of NO_3^-, $SO_4^=$, H^+, Ca^{++}, and NH_4^+. In this discussion we neglect trace metals and organic acids because of generally insufficient data for performing spatial interpolation over regional scales (>100 km).

H^+ and NH_4^+ concentration or wet deposition are of interest due to the potential acidifying effects of these ions on foliage and soils. $SO_4^=$ and NO_3^- concentrations are of interest because they usually derive from anthropogenic emission sources (at Northern Hemisphere continential monitoring sites) and because they may be involved in cation leaching from soils or loss of acid neutralizing capacity from lakes (Sullivan et al., 1988). Cation deposition might be of interest because of added buffering to soils or lakes. NO_3^- and K^+ can serve as nutrients for plants. Cl^- has been used as a "conservative" tracer for the effect of evaporation on precipitation concentrations in an analysis of lake chemical budgets (Brakke et al.,1988; Sullivan et al,1988). Some $SO_4^=$ is deposited with cations such as Ca^{++}, Mg^{++}, or Na^+. This seasalt varies over small spatial scales and is not related to hydrogen ion deposition and pH. Therefore, a regional analysis that includes data from coastal sites might need to correct for seasalt contributions (Junge,1963; Rodhe and Granat,1984; Wright,1988).

Either wet deposition or precipitation chemical concentrations may be relevant depending on the ecological effect of interest. For example, foliar leaching in spruce needles or lake chemistry in watersheds with thin soils might be related to precipitation concentrations while soil buffering processes might respond to wet deposition. Dry deposition (Hicks et al.,1987) or cloudwater interception (Lovett et al.,1982) can contribute substantial chemical inputs to terrestrial ecosystems, especially at high elevations in the eastern U.S.A. (Mohnen,1988; Lindberg et al.,1988). Here we only consider wet deposition (precipitation) because it is the best understood and most intensively monitored of the three deposition pathways.

1.2 Literature review

Junge (1963) operated the first U.S.A. precipitation chemistry network and published maps of ion concentrations including seasalt corrections for SO_4 and several cations. Granat (1974; 1975; 1978; 1986; 1988) discussed errors in areal estimates of wet deposition, network design, spatial variability as a function of intersite separation distance and mapped precipitation chemistry in Sweden. Abrahamsen et al. (1976) published early precipitation chemistry maps for Norway. A number of spatial analyses on U.S.A. precipitation chemistry data have been published including: Semonin (1981), Cowling (1982), Calvert et al. (1983), Munger and Eisenreich (1983), Barrie and Hales (1984) and NADP (1987). Cahill et al.(1985) mapped the spatial distribution of ambient aerosol concentrations for the southwestern U.S.A.

Many recent spatial analyses of precipitation chemistry data describe their selection of either $1/R^2$ weighting, kriging, or subjective interpolation schemes. Husar (1988) and Ellis et al. (1983) used an $1/R^2$ weighting for producing U.S. maps while Nodop (1987) adopts a different objective weighting scheme (previously used by Cressman,1960) for mapping concentrations over Europe. Recent subjective spatial analyses of precipitation chemistry include Pacayna (1987), and Granat (1986) for producing maps for the U.S.A., northwestern Europe, and Sweden, respectively. Knapp et al.(1987) produced maps of U.S. precipitation chemistry without contouring the data values. This avoids misrepresenting spatial aspects in the data but probably does not communicate key features as readily as contours could.

The family of geostatistical techniques referred to as kriging is gaining increasing attention for spatial interpolation. Eynon and Switzer (1983) and Finkelstein (1984) published early applications of kriging to precipitation chemistry data. Seilkop and Finkelstein (1987), Barchet (1987), Wampler and Olsen (1987), Guertin et al. (1988) and Haas et al. (1988) applied kriging to U.S.A. precipitation chemical concentrations and/or wet deposition data. Venkatram (1988) combined kriging with deterministic modeling. Halvorsen et al.(1989) compared two forms of kriging to a subjective analysis of precipitation SO_4 concentrations in Sweden. Bilonick (1985) and Le and Petkau (1988) applied spatial time series models to precipitation chemistry in the U.S.A. Robertson and Wilson (1985) discuss spatial analyses used for design of the NTN network. Lefohn et al. (1987;1988) used kriging to estimate ozone exposures while Simpson (1985) applied kriging for mapping trace metals in soil. Halvorsen and Strand (1987) mapped SO_4 aerosol concentrations over northern Europe using kriging.

This report discusses issues involved in spatial interpolation of precipitation chemical composition data but does not present contoured data; we recommend that reports and published work such as Wampler and Olsen (1987), Barchet (1987), Olsen and Watson (1984), Olsen and Slavich (1986), Seilkop and Finklestein (1987), and NADP (1987) be referred to if contour maps for various years are desired.

2 Data selection

The choice of what data to use may represent a compromise between the need for spatial density, coverage of appropriate time intervals, and data quality. In any case the data selection will be a function of the objectives of the study and the data quality and quantity. A number of data sets exist for precipitation amount and chemical composition complicating what might initially appear to be a simple choice. For regional analysis, a fairly dense network of sites is desireable in order to reproduce the spatial variability of the variable of interest.

2.1 Temporal resolution

Most spatial interpolation of precipitation chemistry data has been performed for annual or longer time periods (Calvert et al. 1983; Barrie and Hales, 1984; Finklestein,1984; Wampler and Olsen,1987; Granat,1986; Seilkop

and Finklestein,1987; NADP,1987; Halvorsen et al.,1989). Data for shorter time periods have been interpolated over regional scales, but less frequently. Haas et al. (1988) interpolated winter wet deposition data, Husar (1988) interpolated monthly data for rain depth and concentration, and Halvorsen and Strand (1987) interpolated seasonal aerosol concentrations over northern Europe.

Shorter time periods present additional problems with data completeness criteria; how many samples should be allowed to be missing in a monthly summary period ? It is likely that data screening for chemical outliers is more critical for shorter time averaging periods because the summaries would be more sensitive to a single inadvertantly contaminated sample (by soil, for example).

The shortest appropriate time period for regional analysis could be addressed as follows: perform spatial interpolation on increasingly shorter time scales until spatial correlation cannot be clearly seen in the data (in kriging this corresponds to an experimental semi-variogram where no clear relationship exists between the semi-variance and lag distance, see below). If the data are stationary over time one can pool the variograms for individual months into a composite variogram. Halvorsen et al.(1989) investigated the spatial coherence for precipitation SO_4 for 29 sites across Sweden and found that annual and four year means could be modeled using variograms, but that monthly values displayed too much scatter to be modeled individually or as a composite variogram (plots of variance versus distance displayed no structure).

Spatial time series (Le and Petkau,1988; Bilonick,1985) can utilize temporally resolved data with the added benefit of identifying sites that are temporally uncorrelated to neighboring sites. For example, one very high altitude site in Switzerland distorted the expected spatial patterns of precipitation sulfate over Europe (Nodop, personal communication) but would have been strongly downweighted by a space-time model due to lack of temporal correlation.

2.2 Precipitation amount data sources

In this paper the term "rain depth" is used interchangeably with "precipitation amount" to refer to the mass flux of water to a horizontal plane (m^3-H_2O per m^2 per year, i.e. m/yr).

The National Climatic Data Center (NCDC) in Asheville,N.C. maintains precipitation records taken by the National Weather Service (NWS) and its cooperating observers representing over 9,000 sites for the United States. Several versions and "cleaner" subsets of these data are available. We define a "cleaner" data subset as being more representitive of actual climatology for the specified time period because unusual samples or sites have been eliminated. This avoids sites with incomplete temporal coverage because of missing observations. These are included below in the order of most to least spatially dense:

1. Hourly raw data are available from NCDC for about 9,000 U.S.A. sites.

2. The densest precipitation amount data routinely requested from NCDC are the "Summary of the Day" (includes hourly precipitation amount) and the "Summary of the Month" for the full 9,000 sites across the U.S.A.

3. The "30 Year Normal" precipitation depth data set contains 'cleaned' monthly and annual averages for the time period from 1951-1980 for about 5,000 sites across the U.S.; latitude and longitude for the sites are provided on a separate data file.

4. An even further 'cleaned' subset of the above data is available from the "Historical Climatological Network" (referred to as HCN or CLIMA) in Oak Ridge,TN. Monthly data for the period 1900-1985 at about 1,200 sites across the U.S. are available. HCN /CLIMA screen NWS data for non-representitive sites and unusual values. This data base represents the highest quality rain depth data set available for regional estimation of wet deposition in our opinion.

5. The National Atmospheric Deposition Program / National Trends Network (NADP/NTN) reports precipitation depth for wet chemistry monitoring sites across the U.S.A. This data source is about a factor of six to ten less dense than the CLIMA/HCN rain depth network (see below for comparisons of CLIMA/HCN and NADP rain depth data). The NADP/NTN data set may be too sparse in some cases for regional estimation of precipitation amount and, therefore, wet deposition (Haas et al.,1988; Husar, 1988).

2.3 Precipitation chemistry data sources

NADP/NTN maintains precipitation chemistry monitoring sites that report weekly values for major ion concentrations. During the period from 1978 to 1980 to 1983 to 1985 the number of sites increased from 22 to 82 to 141 to 195 across the United States. Currently there are over 200 NADP/NTN sites reporting precipitation chemical composition, see Figure 1 for their locations.

Additionally, several other networks exist in the northeastern U.S., southeastern U.S., and in Ontario, Canada including (number of sites): MAP3S (9), UAPSP/OEN (20), CAPMoN (25), APIOS-C (36), APIOS-D (16), EPA-SON (27), and TVA (32). The California Air Resources Board and other states also maintain precipitation chemistry measurement networks. Sweeney and Olsen (1987) describe these networks, their period of record, ions measured, and the location of the sites. Precipitation chemical composition data for these networks is available from the Acid Deposition System (ADS).

Sites that produce spatial discontinuities in mapping should receive special attention. Is there a nearby site with different chemistry ? Can this difference be explained by emission sources or meteorology ? For regional estimation, any rain sample must be representitive of a larger geograpical area to be useful. Unusual sites should represent a smaller "support" area than other sites in a network but if they are included in automated interpolation procedures they may incorrectly influence the results for a larger area.

Figure 1: Locations of NADP/NTN monitoring sites which report weekly precipitation chemical composition. Additional sites are operated by other networks. Lower case letters indentify the locations of individual sites; details are given in NADP (1987).

Reproduced from best available copy.
NADP/NTN MONITORING NETWORK
Locations of active sites in the NADP/NTN network as of 1 July, 1988
0 100 200 300 400 Miles
0 161 322 483 644 Kilometers

2.4 Chemical data preprocessing

2.4.1 Definition of valid samples

As with rain depth data, precipitation chemistry data is available in various versions or subsets, depending on data selection criteria used. Data screening used to eliminate sites or samples of questionable quality or representitiveness should be stated as part of the description of the regionalization procedures. Both NADP/NTN and ADS employ techniques for "cleaning up" the data bases. These may take the form of flagging certain measurements or eliminating questionable samples entirely.

The general ADS approach is that a data set should conform to its own network protocol, consist of only "valid" samples, use sites that have regional representitiveness (a point measurement will be used to characterize a larger region), and meet specified completeness criteria to insure that summary reports are not overly biased by missing data. It is important to carefully choose criteria that identify "bad samples" because the criteria are based on expert opinion and may represent bias (selective inclusion of certain samples) which is traded for improved precision by dropping some samples (i.e.- removing contaminated or unrepresentitve samples smoothes the resulting data distributions). This procedure homogenizes the data; its goal is to select data that reflect the true input from the atmosphere to an ecosystem.

NADP/NTN data summary procedures identify samples that are suspected of being "invalid". A valid sample can be categorized according to the following criteria (NADP,1987): (1) NADP/NTN sampling protocol must be met, (2) the sampling interval must be 6 to 8 days, (3) either the rain gauge depth measurement or the precipitation chemistry sample volume must be available, (4) the sample must be a "wet only" sample type, (5) complete chemistry (all 9 ions) must be available from the lab (i.e., the sample volume must be at least 10 ml.), and (6) the sample is not contaminated (based on the field operator's visual report).

Sweeny and Olsen (1987) describe similar criteria used by the Acid Deposition System (ADS) data base managers to evaluate other networks' data. These include NADP-type criteria with respect to "sample type", sampling period, precipitation amount. "Suspect samples" are declared invalid by ADS. A set of valid sample criteria for networks operating under monthly sampling schedules are added by ADS including (e.g.- for the Canadian APIOS-C network): the sampling time period must be between 21 and 35 days.

Examples of other valid sample criteria that might be applied in addition to those described above include either flagging or removing samples with particular chemical characteristics:

1) sample charge balance deviations (two commonly used formula are: (cations-anions) /(cations +anions) and (cations/anions) where the expected values are 0.0 or 1.0, respectively) provide a measure of the

accuracy and completeness of the chemical analyses for individual samples and for groups of samples (e.g.- frequency distributions of charge balance errors can identify sites with unusual chemistry, Vong et al.,1988),

2) samples with unusually high concentrations of soil-derived species probably are not regionally representitive of atmospheric inputs to terrestrial ecosystems but rather they represent the interaction of precipitation with the local soil cycle (i.e.- site specific dust Husar,1988),

2.4.2 Data completeness criteria

Having decided for each sample whether it is valid or invalid, the next step is to determine if a sufficient number of samples are present to result in a "representative" summary. Several organizations have developed similar criteria:

NADP/NTN evaluates completeness criteria on a seasonal and annual basis and prepares data summaries when (NADP,1987): (1) valid rain chemistry samples for 75 percent of the summary period, (2) for at least 90 percent of the summary period there must be valid precipitation amount from the rain gauge or sample volume (includes periods with zero rain depth), (3) there must be valid samples for at least 75 percent of the total precipitation amount for the summary period, and (4) for the entire summary period the total "sample" precipitation amount (based on conversion from sample volume and collector surface area) must be at least 75 percent of the total precipitation calculated from the rain gauge.

As an example of the impact that valid sample criteria have on data annual summaries, Gibson (workshop presentation) provided the following summary for 1986: 1,695 NADP/NTN precipitation chemistry samples (17 percent of the samples and 15 percent of the total precipitation) were eliminated and flagged as missing in one NADP/NTN data report (NADP,1987) due to following causes:

Reason for Invalidation:	% of invalid samples	% of invalid precipitation
contaminated	32	18
bulk type sample	30	48
inadequate volume	12	4
protocol violated	13	15
sampling period irreg.	9	13
no sample submitted	4	1
lab protocol violated	< 1	< 1

Bias associated with the elimination of contaminated samples applies to under 3 percent of the total precipitation, bias associated with protocol violations applies to about 2 percent of precipitation, and bias associated with inadequate sample volume for chemical analysis applies

to under one percent of the incident precipitation. Only about 6 percent of incident precipitation is lost due to these three important sources of uncertainty during 1986.

It should be noted that better estimates for the northeastern U.S. have been obtained when the Canadian precipitation chemistry data were used in addition to U.S. data (Wampler and Olsen,1987). Besides providing additional data points in the NE region, the Canadian sites have a high station density in Ontario, permitting a better characterization of the spatial covariance at small intersite distances (see below).

2.5 Stratification for regional analysis

The preliminary analysis of any data set is as important as any subsequent modeling. In this section we discuss methods to organize data for spatial interpolation in order to minimize unwanted sources of variability. If the data vary by region or in time they must be properly classified or stratified on that basis. Examples of factors which may influence region to region differences are: meteorology, emission sources, and geology.

There are problems with constructing a single map of precipitation concentrations or wet deposition for an area as diverse as the U.S.A. Termed "first or second order non-stationarity", the problems are that the estimated value and variance may depend on location. First order non-stationarity where the expected mean varies spatially, i.e.- spatial trend (termed "drift" in this manuscript), complicates interpolation processes and the estimation of the uncertainty in the results (see below). Further complicating the situation is second-order non-stationarity where the variance depends upon location.

Regional differences can be dealt with by stratification into more homogeneous sub-groups of data. For example, Haas et al. (1988) divided the U.S.A. into 4 sub-regions before spatial analysis of precipitation chemistry. Cressie (1985) notes that "the choice of regions, disjoint or moving, often is not easy to make".

The basis for such sub-division should be objective. Natural discontinuities such as pollutant sources, mountain ranges, or weather patterns are good candidates for stratification. A clear example is the western U.S.A. where very different topography, meteorology, and chemically different source influences may result in different spatial dependencies than in the northeastern U.S.A. Political boundaries may not provide objective bases for sub-division except where the availability of data is a function of border locations. Since any data stratification provides the basis for subsequent analyses, all data interpretation should include the rationale for the original data stratification, especially any comparison of the sub-regions.

2.6 Comparison of predictions from different data

Bernert and Cusimano (1988) compared two sets of interpolated values for precipitation amount at specific lakes in the northeastern U.S.A. and found general agreement (65% of values were within 10%, 90% of values were within 20% agreement and 99% were within 30% agreement for 883 lakes). The largest disagreements between the two sets of predictions were found in the Adirondack Mountains, especially for lakes at high elevation. It was suggested that either the data selection or interpolation methodology was driving these differences.

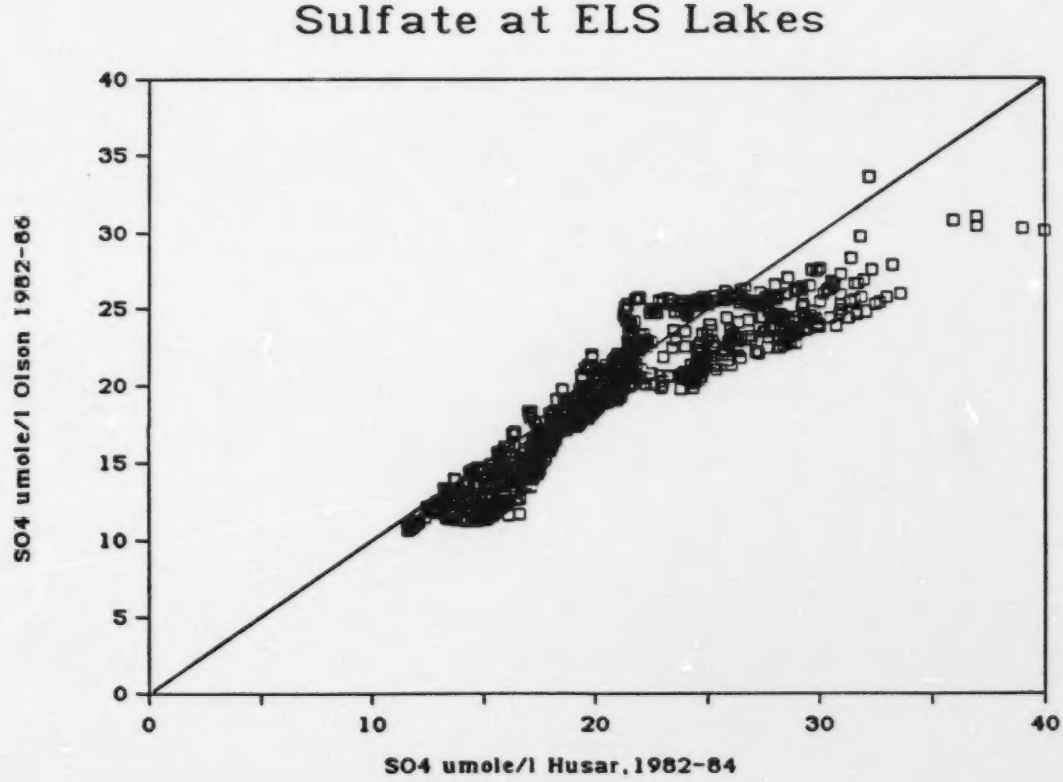
A subsequent comparison of two similar sets of predictions for the concentration and wet deposition of NO_3 and SO_4 were generated by Olsen et al. (1988). Several factors influence the predictions for specific lakes in the northeastern U.S.A. : (1) some difference is associated with the selected temporal period (the inclusion of a high concentration year like 1980 creates differences between the 1980-84 and 1982-86 analyses), (2) different interpolation methodologies were used (ordinary kriging and $1/R^2$ weighting), (3) different spatial density rain depth data were employed (screened "30 year normal" and CLIMA /HCN from 1982-84), and (4) one analysis did not use Canadian precipitation chemistry data.

The results display a fair agreement between the two sets of predictions for both precipitation amount and chemical concentrations (Figure 2). These values were compared according to equation 1:

$$\Delta\ (\%\ \text{difference}) = 100 * (\text{result1} - \text{result2}) / \text{result1} \qquad (1)$$

Most of the percent differences in SO_4 concentration were between 3-13 with many differences below 7 percent. "Result2" predictions were consistently greater than "result1" predictions for all regions. The regions with the worst agreement were: the Adirondacks (18%) and Minnesota (20%). When comparing predicted SO_4 wet deposition it is observed that "result2" predictions were 6-29 percent greater than "result1" with typically nine percent differences. Errors in sulfate and nitrate wet deposition are larger than those for sulfate and nitrate concentrations.

Figure 2: Comparison of two predictions for precipitation sulfate ion concentration where different data selection procedures were employed (Simpson, Husar, and Olsen, workshop presentation).



3 Direct and indirect methods for wet deposition

In generating regional estimates of wet deposition, an investigator might interpolate wet deposition directly or interpolate its two components, chemical concentration and rainfall amount, separately and calculate wet deposition from the product of the two interpolated data fields. Wampler and Olsen (1987) compared these two approaches and obtained slightly different wet deposition results for the same chemical concentration data, especially near sharp gradients in precipitation amount.

3.1 Precipitation amount and concentration variability

Rainfall amount patterns reflect the influence of topography and synoptic meteorology, are spatially variable, and can be mapped using the dense NWS rain gauge network or the less dense NADP/NTN data on precipitation amount. Precipitation amount can be obtained from the NWS at about 10-45 times the spatial density of the precipitation concentration data (in the U.S.A.).

Husar (1988) compared results for interpolating precipitation amount starting with different data sets (using $1/R^2$ weighting, see below). He interpolated monthly data from the spatially dense (721 stations) northeastern U.S. network of rainfall gauges in order to estimate 1984 monthly rainfall amount for 18 "unknown" locations and then compared his interpolated values with independent data (rain gauges located at NADP sites). Results indicated a fair agreement for monthly estimates. He composited the interpolated monthly values into annual average precipitation accumulation; these annual predictions display better agreement with data. Interpolation from the NE network data overpredicts the NADP/NTN precipitation amount. Bigelow (1986) reported a relative mean error of 5.8 percent when comparing three years of NADP/NTN and NWS precipitation depth data for 24 sites across the U.S.A.; he concluded that "rain gauge accuracy has been the most neglected part of the NADP/NTN field operations"

Haas et al. (1988) also found that the spatial scale of the NADP/NTN gauges is larger than the scale over which precipitation amount varies (he couldn't fit variogram models to the NADP precipitation amounts). The NE network for rainfall amount is able to reproduce smaller scale spatial variability missed by NADP/NTN gauges according to Husar's analysis. Lack of station density makes prediction of rainfall amount at non-monitored sites unreliable. The NADP/NTN sites are not representitive for interpolating precipitation amount because they do not capture variability in small scale meteorological processes which drive variation in precipitation amount.

High elevation areas present special problems for spatial analysis because precipitation depths are not well characterized (limited amount of data and problems with collecting snow in exposed locations). Adjusting the estimates of rain depth and/or wet deposition for elevation is difficult because of "rain shadows" on the downwind side of a mountain, and other local effects. Dingman et al. (1988) compared a direct spatial analysis of precipitation amount with a similar analysis of the residuals from the

"altitude-corrected" data in a mountainous region of New Hampshire and Vermont. The altitude corrections provided little or no improvement in their ability to predict rain depth. In general, one is forced into extrapolation rather than interpolation when estimating precipitation amount for high elevation sites.

Correction of precipitation amount data for elevation is difficult over regional scales (>100 km) because of the influence of local factors. Rainbird (1970) suggests that the estimation of precipitation amount for ungauged locations is more satisfactory for means of a large number of storms. Local or point estimates of rain depth might be improved using "corrections" if the investigator has detailed knowledge of the likely controlling factors such as (1) site slope, aspect, and exposure, (2) synoptic meteorology, (3) local wind patterns, and (4) elevation. Therefore, "expert opinion" regarding variation of precipitation amount is recommended for use with high elevation sites only on a case-by-case basis.

Because precipitation concentration data are less spatially dense than rain depth data, the philosophy for mapping wet deposition sometimes has been to interpolate the concentration and precipitation amount fields separately before calculating wet deposition as the product of the two interpolated fields (Wampler and Olsen,1987; Granat,1978; 1988a). We refer to this as the indirect method of interpolating wet deposition. For this approach to produce accurate predictions the two data sources must be representitive of the variability in the processes of interest and concentration and precipitation amount must be independent (or the results must be corrected for the degree of covariation).

Any data field must be continuous for interpolation to be meaningful. Precipitation chemical concentration fields are "smoother" than wet deposition over regional scales (i.e.- they display more spatial coherence), making interpolation more accurate. A smooth concentration field would be consistent with the dispersion and transport of emitted pollutants over broad scales, given that collection sites avoid known local emission sources. Therefore, interpolating precipitation concentration data reflects an emission based process with the resulting predictions for non-monitored locations reflecting the variability associated with emissions, chemical conversion, atmospheric transport, and dispersion.

Bilonick (1985) and Guertin et al. (1988) point out that kriging volume-weighted mean precipitation concentration will produce an 'unweighted interpolation' because the kriging interpolator is an additive operator. This consideration applies mainly where ion concentrations vary over short spatial scales, for example near emission sources. For this case a true volume-weighted mean can be determined as the quotient of separate wet deposition and precipitation amount interpolated fields (effectively a 'volume-weighted -interpolated' concentration). When ion concentration gradients are relatively flat compared to the monitoring site separation, the 'unweighted interpolated' concentration and 'volume-weighted- interpolated'

concentration will be very similar (regardless of variation in precipitation amount, i.e.- the weights vary but the data do not). Additionally, there is no evidence of significant spatial covariance between precipitation amount and concentration (discussed below), making direct kriging estimates of concentration reliable and unbiased (Buxton,1982).

3.2 Direct method for wet deposition

We define wet deposition as the chemical mass flux to a horizontal plane (e.g.- $gm/m^2/yr$) calculated as the product of concentration and precipitation amount. The direct interpolation of wet deposition based only on colocated rain depth and concentration data has several obvious advantages: (1) simplicity in setting up data files, (2) a one step procedure for performing the calculations, and (3) a single set of descriptive statistics to describe the results and their uncertainties. However, Husar (1988) and Haas et al. (1988) found that the variability in rain depth is not captured by the spacing of the NADP sites. This makes direct interpolation of wet deposition data less certain for locations where precipitation amount is different from nearby monitoring sites, a situation frequently encountered in mountainous areas with considerable ecological interest.

Wampler and Olsen (1987) compared direct interpolation of NADP/NTN SO4 wet deposition with separate interpolations of precipitation SO4 concentration and amount (based on a dense NWS grid) followed by calculation of wet deposition (indirect method). Differences exist between the two maps (greatest in the southern Blue Ridge Mountain area) that are linearly related to rain depth. The direct interpolation of wet deposition results in a smaller variation in predicted deposition than obtained from the indirect method.

Seilkop (workshop presentation) reports that interpolating wet deposition directly cuts off the "tails" in the distribution of interpolated values (no extremely high or low values are predicted). Important variability in wet deposition (but probably not precipitation concentration) occurs on a scale smaller than that represented by the NADP/NTN precipitation chemistry monitoring sites. This suggests that the indirect method may be useful because it allows the use of more information on the precipitation amount and, therefore, can better describe the variability in the precipitation process. The direct method requires the use of one parameter whose spatial variation is fairly well described by the data (concentration) but another (precipitation amount for only the NADP sites) that is not completely described for small spatial scales (such as might be needed for prediction at non-monitored locations such as individual watersheds). In the indirect method both the concentration and precipitation amount (using one of the denser NWS data sets) are better described with respect to their spatial variation, resulting in a better description of the spatial variation in wet deposition.

3.3 Evidence supporting the indirect method

Spatially variable precipitation patterns may or may not perturb the smoother precipitation concentration field. It is well known that at a given site the highest concentration events over time are those with low rainfall amounts and vice versa (Farmer et al., 1987; Barrie and Neustadter, 1983). If across sites this "inverse relationship" (negative correlation) held for a single time period, then wet deposition would be overestimated in areas of unusually high rainfall using the indirect method of calculation (when multiplying the interpolated concentration field by the interpolated rainfall amount). However, we present experimental evidence that suggests that concentration and precipitation amount are independent across space.

Table 1: A comparison of changes in annual average precipitation amount and sulfate ion concentration for ten sites (Hubbard Brook,NH, Aurora,NY, Whiteface,NY, Ithaca,NY, Brookhaven,NY, Delaware,OH, Wooster,OH, Leading Ridge,PA, State College,PA and Huntington,NY).

KEY:
X Inverse relationship present
O No inverse relationship observed

YEARS COMPARED:	RESULT:
80-81	X
80-82	
80-83	X
80-84	X
80-85	
81-82	O
81-83	O
81-84	O
81-85	O
82-83	X
82-84	X
82-85	
83-84	X
83-85	X
84-85	

note:
"DRY" YEARS: 80 81 85
"WET" YEARS: 82 83 84

Note: When precipitation amount increased and sulfate concentration decreased (or the converse) between the two years compared, an inverse relationship for annual averages is indicated. When both variables increase or decrease together, no inverse relationship is observed. If the changes are small compared to the annual variability, no information is provided by the comparison.

The analysis of the relationship of annual average precipitation SO_4 concentrations and precipitation amount presented in Table 1 suggests the existence of year-to-year variation in concentrations which are sometimes accompanied by changes in rain depth. One would reach different conclusions regarding concentration/rain depth independence depending upon which years' data were compared, probably because a one year averaging period does not provide a representitve sample of the distribution of rain events. Some years with below normal rain depth had low concentrations (1981) while some had high concentrations (1980) compared to other years (1982-85). Similar results were obtained by Gibson (1988) when comparing annual NADP/NTN data for individual sites located at Kane,PA, Parsons,WV, Caldwell,OH, and Findlay Farm,NC, i.e.- changes in precipitation amount and concentration between years were independent. Of course, these analysis of differences in precipitation concentration and amount for different years do not directly speak to the question of independence across space because different years data contain temporal variability.

Granat (1988a) studied the pure spatial aspects of this relationship by sampling along two transects in southern Sweden for one year. The data from Sweden offer the simplification that most pollutant transport is from the south such that the effect of precipitation amount can be separated more easily than would be possible at locations where transport patterns are more complex. Across the transects the mean precipitation concentrations are nearly constant but "rain depth" varies by almost a factor of two. Figure 3 presents Granat's data for these two transects. Granat concluded that variation in rainfall amount does not alter the spatial patterns in the average rainfall concentration field when the entire study region is influenced by the same "airmass" type. Subsequently Granat (1988b) investigated the precipitation concentration and amount spatial relationship over the Baltic Sea and concluded that direct interpolation of wet deposition for that area overpredicted actual wet deposition. Granat's studies support the use of the indirect method for Sweden.

Lynch and Grimm (1988) monitored precipitation amount and chemical composition along an east-west transect across Pennsylvania. Their data do not support the notion that spatial differences in precipitation amount dominate spatial patterns in the chemical concentration of sulfate ion because both gradients are in the same direction. The complexity of sources and meteorology preclude definite conclusions from the Pennsylvania data but the results are consistent with no spatial correlation between precipitation amount and concentrations.

Figure 3: Annual average precipitation amount and concentration for two transects (A and B) across southern Sweden, as shown in the insert. The X axes represent increasing distance from the west coast of Sweden (Granat, 1988a).

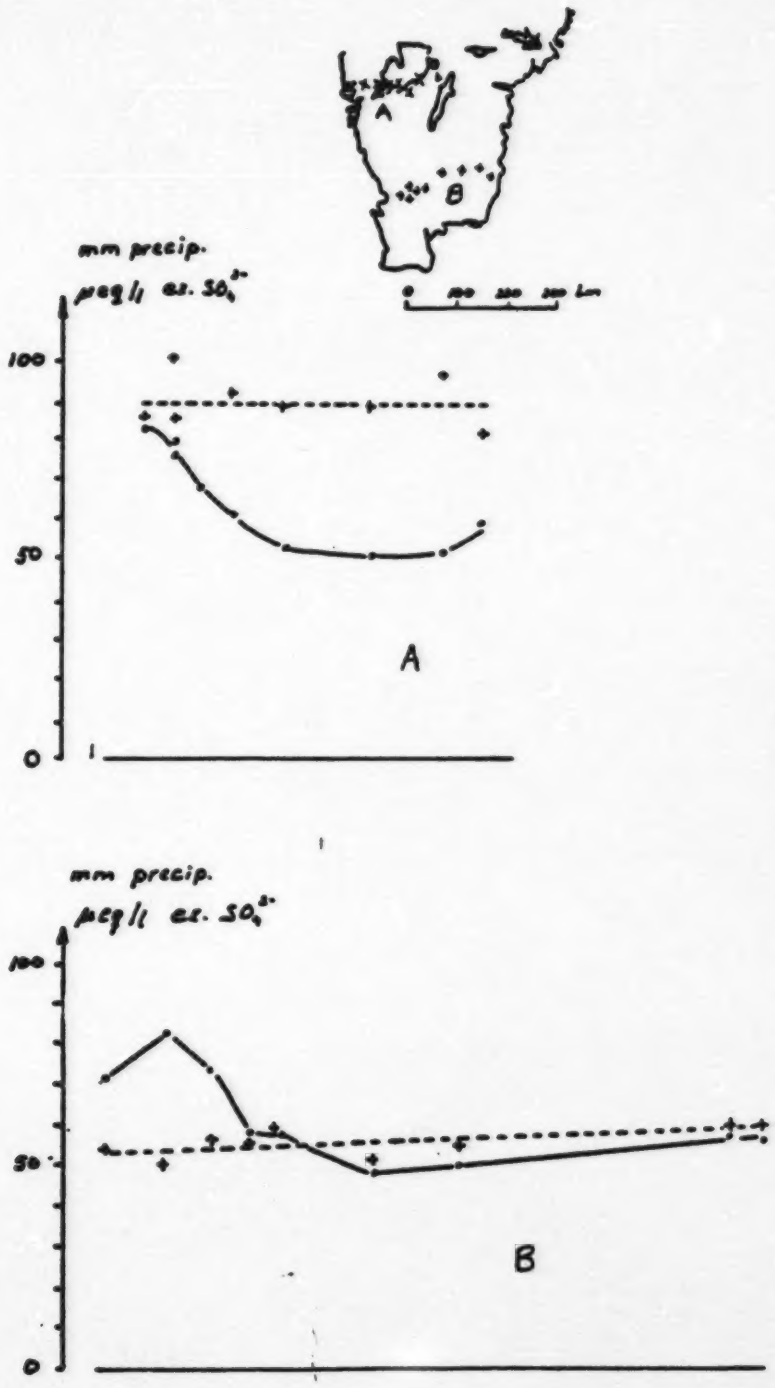
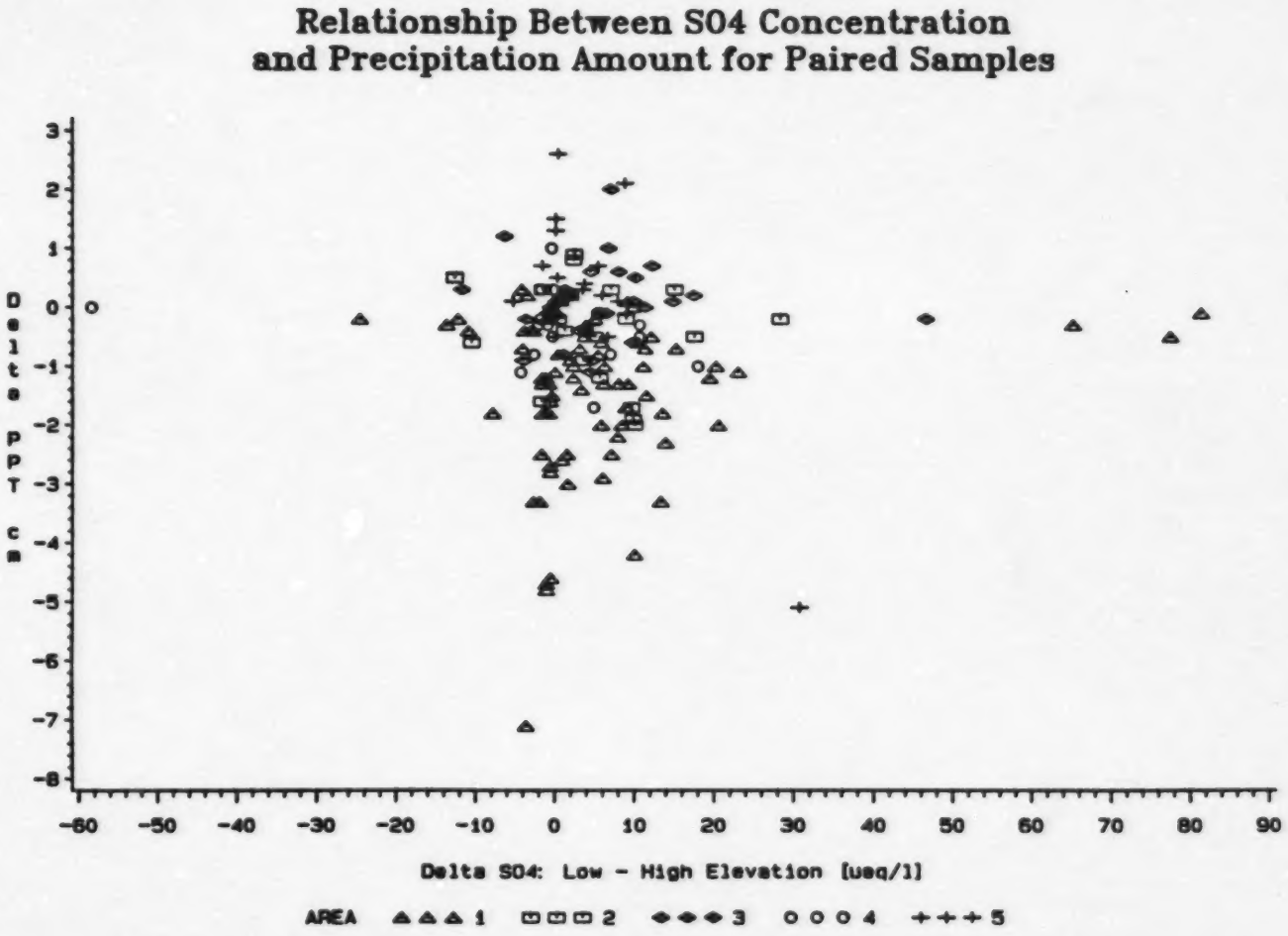


Figure 4: Elevational differences in SO4 concentration (ueq/L) versus elevational differences in precipitation amount (cm) for paired weekly samples collected at five study areas in the Southern Rockies.



NADP/NTN operates paired high/low elevation precipitation chemistry samplers in five airsheds in the southern Rocky Mountains of Colorado and Wyoming (NADP,1987). Each airshed had one sampler located at about 8,000 feet elevation and another at 11,000 feet, see Figure 1 for site locations. Both high and low sites received precipitation predominantly in the form of snow. The low-high differences in SO_4 concentration do not co-occur with differences in precipitation amount (Figure 4) suggesting that some factor other than dilution with liquid water drives the variability in precipitation SO_4 concentration.

O'Loughlin et al. (1986) collected daily samples along a transect from the suburbs of Seattle,WA up to 4,000 feet elevation in the Cascade Mountains. As elevation increased SO_4 concentration always decreased but precipitation amount increased or decreased for different storms. In this study topography may be important in explaining the lower concentrations at high elevation because it provides: 1) a barrier to transport of urban air and 2) lifting and cooling of air which can result in snow in the mountains but rain at lower elevations. This can result in lower SO_4 concentrations in the mountains due to a variation in the precipitation formation mechanism (Scott,1981). Other factors than precipitation amount were important to the spatial variation in precipitation concentrations in the Puget Sound area.

Lindberg et al. (1988) found differences in precipitation amount but not for precipitation chemical concentrations between two sites at different elevations in the Great Smoky Mountains National Park and in Oak Ridge,TN. This suggests that in the absence of data at their high elevation site, the indirect method of determining wet deposition (assuming full data are available for the low site but only precipitation amount for the higher elevation site) would have produced better estimates of wet deposition at the higher site than the direct method (ignoring the high elevation precipitation amount data because of a hypothetical absence of chemical data). They noted that cloudwater deposition and dry deposition further enhanced the differences between the sites such that an increase of a factor of 2 to 5 were observed over an elevation change of 1500 m. in 100 km. Clearly factors other than wet deposition may be very important if spatial patterns in total deposition are desired.

Fowler et al. (1988) collected precipitation along a transect across the Great Dun Fell, a 900 m. isolated hill in Great Britain that frequently experiences orographic cloud. On the average they found that both precipitation amount and concentration increased with elevation with the concentration increase only significant above 600 m. elevation. They concluded that predicting deposition on hills from lower elevation data produced serious error for cap clouds but they were uncertain whether similar differences would exist in other locations. This highlights the importance of understanding site specific factors before charting the spatial variation in precipitation composition. The use of the indirect method is recommended for charting regional scale (>100 km) features in deposition patterns; these regional scale features may or may not be representitive on local

scales depending upon site specific influences. Note that even at great Dun Fell the indirect method would have produced estimates at least as good as the direct method would have produced (albeit both are poor).

3.4 Indirect method for wet deposition

The seeming paradox of a precipitation concentration and amount inverse relationship **across time** but independence **across space** requires an understanding of the governing meteorological and chemical factors. The variation in precipitation amount between events would be related to available moisture, temperature, synoptic scale and local wind patterns, updraft velocity in clouds, and condensation nuclei number concentration. The variation in precipitation concentration between events would be related to upwind emission source strengths, oxidant availability, temperature, aerosol and gas scavenging efficiencies, and atmospheric ventilation. The inverse relationship between amount and concentration **over time** at one location is probably related to scavenging efficiency changes from one event to another, i.e.- precipitation formation mechanism or dilution of pre-existing aerosol with varying liquid water (Scott,1981; Barrie and Neustader,1983). **Across space** the scavenging efficiencies for a single mixture of events would not be particularly variable except where sharp topographical features dominate air flow. The point is that spatial independence of concentration and amount is related to **within** airmass/cloud variations while the well known inverse relationship across events is likely driven by the difference **between** airmass/cloud systems.

We have reviewed seven experiments that reveal general concentration and precipitation amount spatial independence; this suggests that factors other than rain depth control spatial variation in precipitation concentrations. Two comparisons of annual means, although not directly testing the spatial aspects, are consistent with this in that no consistent inverse relationship was demonstrated. For many cases, it appears reasonable to interpolate available precipitation concentration data and convert to local wet deposition fluxes using a more densely sampled precipitation depth data base.

Further analyses of the independence of concentration and rain depth across space are recommended for individual regions to determine whether site specific factors may control spatial variability in these two parameters and their degree of covariation. For example, if precipitation amount and pollution emissions had gradients directed in opposite directions due to source location and wind direction control, an inverse relationship might be observed.

One criterion for preferring either direct or indirect estimation of wet deposition is the need to minimize the bias and uncertainty in the results. By this criterion, if the propagated error for the multiplication of the concentration and rain depth is less than the interpolation variance for

direct analysis of wet deposition alone, the indirect method might be preferred. To date this comparison has been performed once (Seilkop, workshop presentation) and showed similar but slightly smaller errors for the indirect approach.

It is possible to evaluate in a statistical sense the appropriateness of the independence assumption and, therefore, the error associated with the indirect method of estimating wet deposition for individual data sets. If independence does not hold, the wet deposition will be biased in proportion to the correlation across space for rain depth and concentration. Rigorous testing for independence is difficult in this case because the distributions for each random variable are likely to be site dependent. Converting the concentration and amount data to Z-scores (subtract the site mean and divide by the site standard deviation) would allow a test of correlation if one assumed that each process had the same family of distributions over the region of interest.

Annual average SO_4 wet deposition was relatively constant for the same 1980-85 time period reported in Table 1; errors would be made in estimating wet deposition from separate analyses of concentration and rain depth if different time periods are used for the two pieces of data. For example, if the "30 year normal" rain depth for the U.S. was matched with SO_4 concentration from 1980, the resulting deposition would not be a good estimate for either 1980 or any longer time averaging period. It is important that indirect method for estimating wet deposition utilize data for corresponding temporal periods.

4 Spatial interpolation techniques

Spatial interpolation is used for generating maps and for estimating data for non-monitored locations. Interpolation revolves around this question: "what weights should be assigned to nearby (geographically oriented) data in the estimation of some unknown (interior) point ?" Intuitively, points close to estimated point should receive more weight than distant points, but how much more ?

4.1 Overview of interpolation approaches

Interpolation methodologies have been used for generation of isopleths for about five decades. Initially in the 1920's cartographers generated isopleth maps by visually interpolating point values on a mapped surface by generating lines of equal magnitude. This is referred to here as a "subjective" methodology. With the advent of modern computers, quantitative methods for interpolating surfaces were used to generate isopleths. Burrough (1986) suggests classifying these methods into two categories: deterministic and stochastic, see Table 2.

Table 2:

A comparison of interpolation methods (Burrough,1986).

A comparison of methods of interpolation

Method	Deterministic/ stochastic	Local/ global	Transitions abrupt/ gradual	Exact interpolator	Limitations of the procedure	Best for	Output data structure	Computing load	Assumptions of interpolation model
"Eyeball"	Subjective/ deterministic	Global	Abrupt	No	Non-reproducible, subjective	Field data, aerial photo interpretation	Polygons	None	Intuitive understanding of spatial processes; homogeneity within boundaries
Edge-finding algorithm	Deterministic	Global	Abrupt	No	Often requires shapes to be defined and stored; better for man-made features than for natural landscapes	Raster images from remote sensors	Raster	Moderate	Homogeneity within boundaries
Proximal (Thiessen poly.)	Deterministic	Local	Abrupt	Yes	One data point per cell; no error estimates possible; tesselation pattern depends on data point distribution	Nominal data from point patterns	Polygons	Light/ moderate	'nearest neighbour' gives best information
Trend surface	Stochastic	Global	Gradual	No	Edge effects, outliers, complex polynomials do not necessarily have meaning; errors are rarely spatially independent.	Demonstrating broad features and removing them prior to other methods of interpolation	Points on a raster	Light/ moderate	Multiple regression—phenomenological explanation of trend surface; independent Gaussian errors
Fourier series	Stochastic	Global	Gradual	No	Not applicable to data; lacking periodicity	Periodic features such as sand dunes, ripple marks or gilgai, or man-made features	Points on a raster	Moderate	Strict periodicity in phenomenon of interest.
B-splines	Deterministic	Local	Gradual	Yes	No estimates of errors; masks all uncertainties in surface	Very smooth surfaces	Points on a raster	Light/ moderate	Absolute smoothness of variation
Moving average	Deterministic	Local	Gradual	No unless constrained	Results depend on configuration of data points and size of window; simple versions assume isotropy; no error estimates unless retrospectively calculated	Quick contour plots of moderately smooth data.	Points on a raster	Moderate	Continuous, differentiable surface is appropriate
Optimal interpolation (kriging)	Stochastic	Local	Gradual	Yes	Practical and theoretical problems of non-stationarity in data; large computing costs for mapping	Situations where the most detailed estimates and their errors are required	Points on a raster	Heavy (very heavy for universal kriging)	Intrinsic hypothesis (homogeneity of first differences); average local values can be represented by a continuous surface.

Deterministic methodologies were applied to fitting surfaces in the field of numerical analysis. Deterministic spatial interpolation approaches fit the data exactly, implying that no sampling or estimation error exists. Linear interpolation initially provided prediction of values but this algorithm sometimes provided poor estimates. Weighting schemes for individual point values became incorporated into spatial interpolation methods. Inverse distance weighting is a method to generate estimates as a function of distance. Other deterministic methods such as splines have recently been introduced (splines involve fitting localized quadratic terms to data and successively recalculating the quadratic for moving regions across the area of analysis). The data analyst using deterministic methods must choose *a priori* the exact functional form for weighting observations or fitting surfaces. This approach is useful when the factors governing spatial variability are well known.

Stochastic methods approximate the data based on statistical measures about the surface. In the 1950's trend surface analysis was applied to geographic data. A trend surface is similar to a regression through space where the data are a function of the spatial location. In effect, trend surfaces represent the "best estimate" from a smooth surface. Nonlinear transformation or polynominals have been used to define simplistic surfaces. This approach defines an overall trend and predicts data values which have corresponding residuals. Fourier series have also been used for stochastic models which possess periodicity. Kriging is a relatively recent interpolation methodology which predicts values and associated error measures.

These quantitative methods are often referred to as "objective". In fact they each are partly subjective in that decisions as to what is to be optimized by the technique are involved. For example, kriging minimizes the estimation variance while splines fit isopleths that correspond to a "minimum energy" in an analogy to curvature in a stiff wire. The automatic, quantitative techniques probably are referred to as "objective" because they are systematic in the way that assumptions are applied to data.

4.2 $1/R^2$ weightings

Among objective interpolation schemes, 'one over distance squared' weighting has been popular. However, no published information suggests that this exact functional form of weighting samples is known to be appropriate for charting precipitation chemistry spatial variation. The "$1/R^2$" (where R is the radial, geographic distance from a known data point to the point to be estimated) scheme downweights distant points relative to near sites more than inverse distance weighting does. This method produces a weighted average of nearby data; $1/R^2$ approaches do not determine a functionally explicit trend but rather smooth the input data. Buxton (1982) concluded that $1/R^2$ estimates of coal reserves were less precise than kriging estimates using the same number of data.

Husar (1988) used 12 nearby sites (if they were within 500 km of the point to be estimated) to produce $1/R^2$ weighted averages. This technique weights each of several closely spaced sites as much as a single site at the same distance in an opposite direction from a point to be estimated. A comparable kriging analysis would downweight each of the closely spaced sites relative to the single site; the clustered sites would be treated as somewhat redundant information. Since NADP/NTN sites are relatively evenly spaced, this methodological difference should not dominate the results.

One interesting note is that inverse distance squared ($1/R^2$) weighting can be expressed in terms of a particular variogram model. The theoretical variogram model which corresponds to $1/R^2$ weighting passes through negative variances, an undefined condition mathematically (Henning Omre, personal communication). Inverse distance ($1/R$) weighting can not be equated to any variogram model.

Uncertainty estimates for $1/R^2$ weighted predictions could be generated via a cross-validation approach (i.e.- leave out a data point and interpolate a value from the rest of the data) as was done by Buxton (1982). Also, an experimental variogram could be used to estimate uncertainties for $1/R^2$ weighting; variances would be pooled from the variogram depending upon the configuration of points used in each estimate. Not estimated is the bias associated with this weighting scheme making estimation of confidence intervals problematic. Uncertainty estimates have not been provided for precipitation chemistry using techniques other than kriging, possibly because software containing these techniques have not routinely incorporated such features.

4.3 Subjective analyses

Subjective analyses utilize the analyst's judgement as to what weight to apply to surrounding data. Essentially, these analyses involve the use of expert opinion. In practice, subjective analyses can utilize sophisticated human algorithms for determining weights for surrounding data points and are capable of producing maps that accurately model spatially oriented data. Granat (1986) constructed contours for precipitation NO_3^- and $SO_4(xs)^=$ in Sweden by considering the location of emission sources, local and regional climatology, collection site characteristics, and the dependence of atmospheric chemistry on air trajectories. It is acknowledged, however, that different individuals produce different maps for a single data set. At this time no one has reduced the necessary chemistry, meteorology, and other factors to a computer code capable of interpolating precipitation chemistry data with accuracy and precision.

A comparison of Granat's map with independent kriging estimates suggests that his map performed as well as the kriging procedure did in terms of how well map isolines describe the data (Halvorsen et al.,1989). Not estimated was how well his maps would predict data that were unknown (as done with cross validation approaches). Note that Sweden probably presents a simpler system than the eastern U.S.A. for such a regional analysis because most

pollutants are transported from the south towards Sweden. Granat's subjective approach did not estimate the uncertainty associated with his interpolation procedures.

4.4 Kriging

Kriging is the spatial interpolation technique used in "geostatistics". A good background to this can be obtained from: Matheron (1971), Journel and Huijbregts (1978), and Clark (1979). Kriging is a system of simultaneous equations whose solution produces interpolation weights that are based on a model of the covariance structure in the data. Kriging provides local estimates of data values for non-monitored locations and values of the estimation variance. Tabios and Salas (1985) compared several techniques for spatial interpolation of precipitation amount and concluded that kriging and "optimal interpolation" techniques were superior to the other techniques for the data sets they analyzed.

4.4.1 Assumptions.

Kriging produces interpolated values for non-monitored coordinates in generating regular, rectangular gridded data necessary for contour maps. The predicted value for an unknown point is the weighted average of the other values. The spirit of kriging is that the relative interpolation weights for near and distant sites are based on the degree of covariance in the measured data. A variogram model describes this spatial covariance structure.

Although kriging or any other statistical algorithm will generate estimates, these estimates are meaningful only to the degree that the underlying assumptions are satisfied. In kriging the assumptions are that the true semi-variogram is known and the process is second order stationary (the variogram is constant over the region; see also "stratification" above). The interpolated values and the variance estimates are the "best linear unbiased estimates (BLUE)". Armstrong and Jabin (1981) point out that any variogram model must be positive definite to avoid negative variance estimates.

In ordinary kriging the expected value for any point is the data mean; this assumption is termed "first order stationarity" and implies that this mean is not changing over the region. Ordinary kriging assumes first and second order stationarity. When any drift is present universal kriging can be used and it is assumed that this drift can be modeled using a polynomial form. Data need not be isotropic (defined as "having no directional dependence") for either form of kriging but anisotropic data orientations must be reflected in the variogram modeling to obtain representitive variance estimates.

4.4.2 Variogram modeling

Kriging assigns weights based on a model of the semi-variogram. In generating a variogram, one accumulates pairs of measurements according to the separation distance (lag distance or "lag") between the sites. The

interval size is chosen to maximize resolution in the variogram while accumulating at least 30-40 observation pairs per lag (in order to accurately characterize the variance). Class intervals based on distances are used as bins; the intersite variances are averaged for each lag distance interval. Figure 5 presents a plot of an experimental semi-variogram as: the lag ("h", X-axis) versus the average of the squared data differences ("gamma", Y-axis) for all pairs of sites within the lag interval. The experimental variogram is calculated as:

$$\gamma(h) = 1/\ 2*N(h) * \Sigma\ \Sigma\ (X_i - X_j)^2 \qquad (2)$$

where: X_i, X_j are the concentration or deposition in the h^{th} distance bin
$\gamma(h)$ is the semi-variogram estimate for the h^{th} distance bin
$N(h)$ is the number of pairs of data in the h^{th} distance bin
for: $i = 1, N$
$j = i+1, N$

In previous applications to environmental data (see above), there has been subjectivity involved in choosing a variogram model given an experimental semi-variogram. This "variogram model" takes one of four basic forms (linear, exponential, spherical, gaussian) to allow solution of the kriging equations. Equations 3 and 4 present spherical and exponential variogram models, respectively (see Clark (1979) or Journel and Huijbregts (1978) for the linear or gaussian variogram models):

<u>The spherical variogram model</u>:

$$\gamma(h) = C * (3h/2a - h^3/2a^3) + C_o \quad \text{for } h < a \qquad (3)$$
$$= C * C_o \quad \text{for } h > a$$

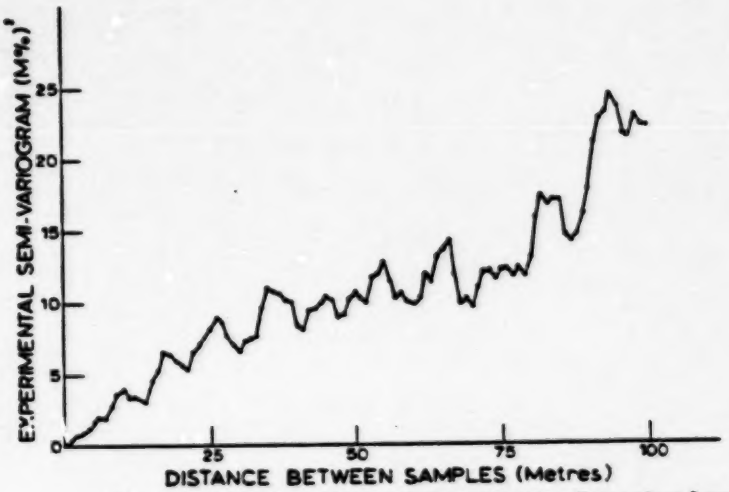
<u>The exponential variogram model</u>:

$$\gamma(h) = C * (1 - \exp(-h/a)) + C_o \qquad (4)$$

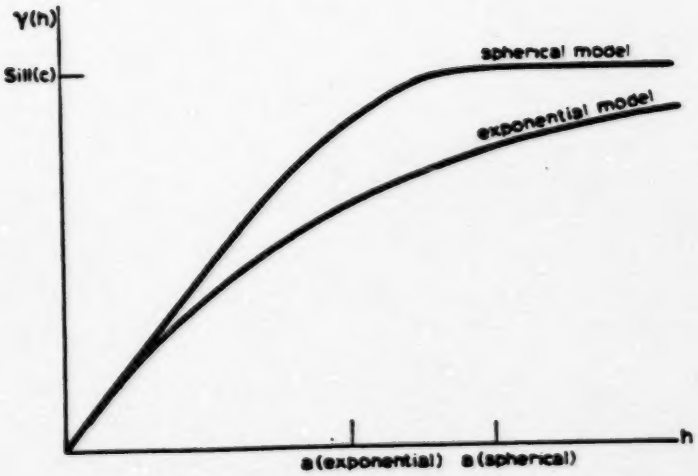
where: a = range
C = sill
h = lag distance
$\gamma(h)$ = semi-variance
C_o = nugget

All forms of the variogram model are characterized by a few important parameters: range (lag distance at which the semi-variance stops increasing), sill (the value of the semi-variance at the range which is the population variance), and nugget (semi-variance at zero lag distance), see Figure 5.

Figure 5: Examples of experimental semi-variogram and two frequently used variogram models



Experimental semi-variogram constructed on the silver values from a complex sulphide deposit.



Comparison of the exponential and spherical models

The nugget is an important parameter in the variogram model because it serves to smooth or dampen the final isopleths when it is relatively large compared to the population variance (Buxton,1982). The nugget is the minimum variance that the data can address. Finklestein (1984) investigated the nugget for precipitation chemistry data in the eastern U.S.A. and compared the results to co-located NADP samplers; he concluded that the nugget was similar in magnitude to measurement error. Many kriging applications take the approach that the spatial variability at very small scales is the measurement error.

The slope of the variogram model near the origin (at the same lag distance as the closest spacing of sites) is important because it determines relative weighting for the most influential data points in a kriging analysis. The semi-variance at small spatial scales is usually not well defined by the data because few precipitation chemistry sampling sites are located at short separation distances (Note: the Canadian APIOS network does characterize short spatial scales). The "sill" in the variogram model describes the semi-variance at large lag distances. The sill describes areas where no spatial dependence is evident and, therefore, represents the population variance.

The final data maps produced by kriging are not particularly sensitive to the exact form of the variogram model but that the kriging standard deviation is sensitive to several features of the variogram (Buxton,1982; Halvorsen et al.,1989). Although not previously applied to spatial analysis of wet deposition, influential point analysis and weighted regression could be used to determine the best variogram model, given an experimental variogram (Cressie, 1985; Hawkins and Cressie,1984). Burrough (1983) describes interpretation of variograms.

If the degree of fit between the experimental semi-variogram and the variogram model is relatively poor, the estimated weights and interpolation variance are not meaningful. In that case *a priori* weighting might be more appropriate than a variogram model which cannot detect any spatial covariance in the data.

4.4.3 Interpolation

Once the variogram model is selected, the kriging weights are defined as a function of the orientation and distance between sites. The prediction (interpolation) for any non-monitored point is a weighted average of the monitored values. Very distant points are so downweighted that they contribute little to the predicted value unless the variogram has a relatively large nugget.

When the data are stratified, separate variogram models are used for each sub-region. When interpolating values for non-monitored points near the boundaries between sub-regions, Haas et al. (1988) used data from sites in adjoining sub-regions for prediction.

Much of the local roughness in a kriging solution goes unobserved when producing a contour map because (1) the predicted values "recover" from spikes near monitoring sites quickly if the nugget is non-zero and (2) grid intersections rarely fall **exactly** at a monitoring location. A technique referred to as "block kriging" estimates average values for several adjoining grid squares such that no spikes ever occur due to the properties of the technique itself (Englund and Sparks,1988).

4.4.4 Drift.

The first order stationarity condition is not met when drift (spatial trend) exists in the data because the expected value is not constant over the region. Because most kriging applications do not include distant points in the estimation procedure (they would receive very small weights and not influence the interpolated value), only the points in the "radius of influence" must conform to the stationarity assumption, i.e.- "local stationarity" must hold but global stationarity is not required.

If variogram models are fitted to data with drift, the variance will be overestimated at large lag intervals unless a "drift free" variogram is calculated (Seilkop,1983). Cressie (1986) suggested the use of "median polish" for handling drift affected data. Alternatively, an experimental "relative variogram" can be calculated (Englund and Sparks,1988; Cressie,1985; David et al.,1984) as:

$$\gamma(h) = 1/\ 2*N(h) * \ \Sigma\ \Sigma\ \ \{(X_i - X_j)/X_{mean}\}^2 \qquad (5)$$

where: X_i, X_j are the concentration or deposition in the h^{th} distance bin

$X_{mean} = (X_i + X_j)/\ 2.$

$\gamma(h)$ is the semi-variogram estimate for the h^{th} distance bin

$N(h)$ is the number of pairs of data in the h^{th} distance bin

for: $i = 1, N$

$j = i+1, N$

The relative variogram produces a "nugget" that is analogous to a coefficient of variation, i.e.- experimental errors are proportional to the measurement value (in ordinary kriging the nugget corresponds to absolute measurement error). This approach tends to reduce the effect of drift on the variogram because data variances near any deposition maxima (such as the Ohio Valley) are scaled down compared to variances in areas with lower wet deposition. Cressie (1985) shows that using relative

variograms on untransformed data is similar to using "standard" variograms with log transformed data. David et al. (1984) concluded that relative variograms reduce the conditional bias associated with kriging highly skewed data.

Universal kriging was conceived for handling 'drift affected' data and has been applied to precipitation chemistry and amount. In this method a variogram model and the drift must be simultaneous estimated, difficult unless the form of the drift is known. In universal kriging, each estimated value has a determinstic component and a stochastic component (interpolated data value = (local estimate based on the drift + kriging estimate from the drift corrected residuals)).

Seilkop (1983) compared ordinary and universal kriging on synthetic data with nonlinear drift and found that universal kriging gave only slightly more accurate predictions. Seilkop and Cohn (1987) investigated generalized covariance kriging for drift affected data and concluded that was ineffective because it oversmoothed data.

Halvorsen et al. (1989) obtained similar results for universal and ordinary kriging for mapping precipitation sulfate concentrations over Sweden. In this case a linear drift was acceptable. No substantial improvement in predicted values or uncertainties was obtained by accounting for the drift when compared to using ordinary kriging (a radius of influence reduced the drift effect).

There appears to be few benefits using universal kriging rather than ordinary kriging while considerable additional computational complexity is needed for universal kriging. We recommend that anyone who is not thoroughly familiar with handling drift in a kriging analysis utilize the following approach:

1) carefully define an experimental semi-variogram and a variogram model with an appropriate "sill",

2) utilize ordinary kriging with a radius of influence small enough to keep the drift effect as mild as possible and

3) interprete the variance estimates as being inflated due to the drift.

4.4.5 Uncertainties

The kriging solution corresponds to the set of weights that minimizes the interpolation variance. The largest kriging variances occur for areas with the lowest data density. The maximum kriging standard deviation is a function of the "sill" of the semi-variogram (i.e. the variance of the data set) and the number of points used in the estimate. The predicted kriging estimation variances will approach the value of the sill divided by N in areas of low sampling density. The lower limit of

the kriging standard deviation is governed by the "nugget". Therefore, the form of the variogram model which is fit to the data will influence the kriging standard deviation map.

An understanding of drift, local maxima or minima, and the radius of influence used in kriging are necessary to interprete the validity of the kriging standard deviation. In the presence of drift or other non-stationarity the kriging standard deviation will be overestimated unless the variogram is "drift free" (Seilkop, 1983).

The kriging standard deviation represents only the "estimation variation". A second uncertainty exists in kriging that is related to the estimation of a variogram model from the experimental semi-variance. Obviously, the better the fit between the variogram model and the experimental semi-variogram, the smaller this second form of uncertainty. This second uncertainty has not been estimated for precipitation chemistry kriging analyses.

Cross-validation (similar to jacknifing) is used to test whether the variogram model and search radius can accurately reproduce the spatial variability of the sampled observations (Englund and Sparks,1988). Cross-validation (CSV) involves estimating values at each location in a sampled area by kriging the neighboring sample values. Unfortunately, the cross-validation residual is a poor estimator of the interpolation error due to a sample size of only one (lacking degrees of freedom). Therefore, one would expect to see a lot of scatter in the CSV residual, making any confidence interval very large for comparison with the kriging standard deviations.

The cross-validation approach is useful for comparing predictions. Seilkop (workshop presentation) compared the the direct and indirect methods for interpolating wet deposition and found slightly smaller CSV residuals using the indirect method. The CSV errors were weakly a function of the magnitude of the prediction. To obtain more information from kriging cross-validation residuals, Haas et al.(1988) normalized them by the kriging standard deviation and plotted the ratios against the site X or Y coordinate to observe any drift in his precipitation chemistry data. Diaconis and Efron (1983) discuss the use of bootstraping for uncertainty estimation.

One approach to determine the accuracy of a spatial analysis would be to predict data for non-monitored sites, monitor for an appropriate time interval, and calculate the true errors. Granat (1988b) used that approach in conjunction with his subjective spatial analyses of wet deposition over Sweden.

If a drift free variogram model can be constructed, the kriging standard deviation probably represents a reasonable estimate for the prediction uncertainty associated with regional estimates of precipitation chemistry. The confidence intervals that can be obtained from kriging variances are only appropriate for symmetric data distributions. This may

not be the case for precipitation concentrations or wet deposition (e.g.- Vong et al.,1988a,b). The use of the kriging uncertainty (with or without a separate experimental error component) is analogous to using the standard normal test as an exact measure when the degrees of freedom are unknown. Caution should be used in stating any confidence intervals from kriging results.

5 Discussion

Both practical and statistical considerations are important for an accurate charting of the regional distribution of wet deposition. Because computerized interpolation (a mathematical/statistical approach) is increasingly used for mapping, statistical constraints must be recognized to achieve unbiased results. However, the available data may not always fully conform to the assumptions that statistical theory may require. However, as in all real world data analyses, the question is not whether some assumption such as "independence" is met, but to what degree the techniques are invalidated if the assumptions are violated.

The choice of the direct or indirect method for calculating wet deposition is based on the variability in precipitation concentration (an emission based process) and precipitation amount (a meteorological based process), the representativeness of available data in reproducing that variability, and in the degree of spatial independence between concentration and precipitation amount. For a smooth concentration field represented by the relatively sparse NADP/NTN data and a more variable precipitation amount field represented by the more dense NWS data, the indirect method probably will produce better regional estimates than directly interpolating wet deposition.

Some interpretation of the processes governing spatially oriented data should be performed to insure that results are not physically unreasonable. For example, if a "variogram model" reveals variation in rainwater acidity over spatial scales that are completely different from what an atmospheric chemist would expect, then the results should be reexamined for deviations from the statistical assumptions or for some misinterpretation of the experimental data.

Although it might seem reasonable that several analyses would produce the same value for SO_4 wet deposition at a non-monitored site, a range of values has been generated by the various investigations referenced herein. Data selection and preprocessing appear to be at least as important as interpolation methodology for explaining differences between two maps (e.g.- Husar,1988; Olsen et al.,1988; Wampler and Olsen,1987). A number of statistical, meteorological, and chemical issues have led scientists to take sometimes divergent approaches to selecting a data set.

The timing of various data analyses may be critical because of temporal variability. Depending upon the appropriate time period for a specific application

(e.g.- deposition to a watershed for a particular water year as contrasted to expected future deposition for a typical year), the data analyst may draw on different data subsets and produce different results.

The comparison of two spatial analyses presented in Figure 2 did not isolate the effect of interpolation methodology from differences attributible to the selection of the precipitation amount and chemistry data sets and subsequent preprocessing steps. An objective comparison of the spatial interpolation methodology might include a cross-validation on both $1/R^2$ weighted predictions and kriging results for the same data set (or other methods that are capable of truly ignoring a subset of data). If data were omitted before the analyses, one validation criterion would be which technique produced estimates closer to the withheld values. A "precision" calculation of interest would involve selection of "random" coordinates at which each technique must predict values. An overlay of contour maps demonstrated that the various interpolation techniques produce similar maps when a common sulfate ion concentration set is used (Bigelow and Stensland,1988; Halvorsen et al.,1989).

6 Summary and Conclusions

In this report we have presented a number of approaches for strengthening any spatial analysis of precipitation chemistry. The goal is that future investigators will benefit from the experiences discussed herein and that they will document key portions of their analyses so as to permit evaluation by peers.

When performing a regional analysis of precipitation chemistry four key issues are: the data selection, data compositing, the interpolation technique, and the uncertainty of the results. Using NADP/NTN and Canadian precipitation chemistry data (at a minimum), screening the chemical data, and using supplemental NWS precipitation amount data appears to present a useful and valid approach to producing regional analyses of concentrations or precipitation amount. The NWS data were seen to reproduce the variability in precipitation amount better than the less spatially dense NADP *precipitation amount* data. The precipitation chemistry data appeared adequate to chart the variation in *chemical concentrations* if the data were first stratified into fairly homogeneous regions. Data selection is critical to the results, more so than we had anticipated.

Direct and indirect methods for charting spatial variation wet deposition were investigated. The indirect method allows the use of more representative precipitation amount data but assumes that precipitation amount and concentration are not spatially correlated. A review of experimental evidence suggests that there is no strong relationship between SO_4 concentration and precipitation amount across space. For most cases, it appears reasonable to interpolate precipitation concentration and convert to local wet deposition fluxes using interpolated rain gauge data. Further analyses of the independence of concentration and precipitation amount *across space* are recommended, especially where site specific factors may control wet deposition.

It is important to understand the rationale for selecting an interpolation technique and the comparability between the technique(s). Given that there are many ways to weight near and distant data for estimating data an non-monitored site, why choose one over another ? One interpolation technique, kriging, determines interpolation weights based on the autocovariance in the data while several other methods assign *a priori* the relative weights (a deterministic approach). Two approaches may produce maps that are similar but estimation variances that are different (or absent). Given that effects researchers want to know the uncertainties in the predictions, kriging is currently a preferred technique. No direct comparison of interpolation techniques with rainwater concentration data was performed.

Regardless of the choice of data and technique, it is important that the statistical, meteorological, and chemical basis for regional analysis be sound. In that regard, it seems likely that interdisciplinary approaches will produce the most useful and accurate regional characterizations of acidic deposition data.

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